Multiphase Equations of State for the SESAME Database

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The great majority of the equations of state (EOS) prepared for the SESAME database until now have not treated the effects of phase transitions except for melting, and then only phenomenologically. We are now developing the techniques necessary to produce for the database a genuine multiphase EOS, in which the thermodynamic properties of the phases are computed individually and the equilibrium phase boundaries are determined by matching Gibbs free energies. We are producing a multiphase EOS for tin as a test case. The large range of compressions and temperatures covered by a typical SESAME EOS ($0 < \rho / \rho_0 < 10^4$ and $0 < T < 10^4$ eV) presents special challenges for such calculations.

The tin EOS contains the beta and gamma solid phases and the liquid phase. The alpha phase is excluded because the beta phase is slow to transform to alpha, rendering beta metastable except for very long-lived processes. To produce this EOS, we first constructed the individual phases, which involved introducing new models into OpenSesame, our new EOS production code. In particular, we needed a model for the ionic contribution in the liquid phase; for this, we used the ionic treatment developed by Chisolm and Wallace in T-1 [1], using at low temperatures the vibration-transit (V-T) theory of liquid dynamics developed recently in our group (see [2] for a review), and interpolating smoothly to an ideal-gas-like expression at very high temperatures. We also incorporated a new treatment of the liquid cold curve due to V-T theory.

To combine the EOS for the different phases and construct the equilibrium phase diagram, we used a treatment inspired by the infinite-time limit of a kinetic model used by Greeff et al. in their work on titanium and zirconium [3]. This method allows the treatment of an arbitrary number of phases without restrictions on the topology of the phase diagram. A difficulty we encountered at this point is the spurious re-emergence of a phase far from where its free energy was calibrated. Our algorithm allows us to restrict a phase to within a window in the ρ – T plane to avoid this problem.

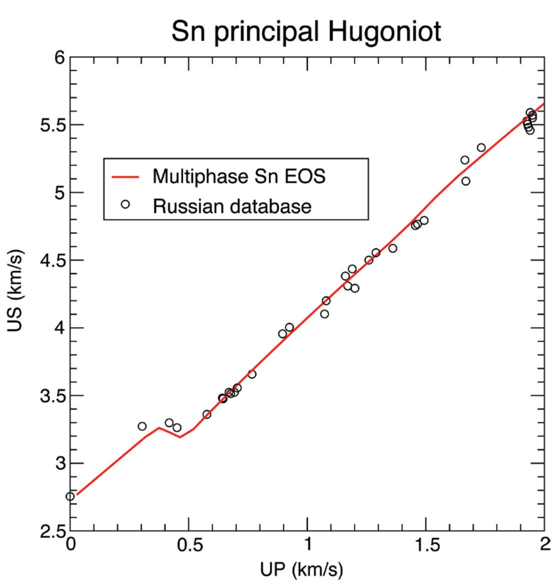

Fig. 1.
The principal Hugoniot of tin predicted by our multiphase EOS compared with experimental data from the Russian shock physics database. Notice that the EOS reproduces the effects of the beta-gamma phase transition as indicated by the data.

An important application of multiphase EOS is to allow for phase-dependent strength models in continuum mechanical simulations. To facilitate this, we are extending the SESAME data format to include phase information. In addition to tabulating the pressure and internal energy as functions of density and temperature, we also tabulate the mass fractions of the phases. This completely specifies phase information in mixed and pure phase regions.

The Hugoniot predicted by this EOS is shown in Fig. 1 with experimental data. The beta-gamma phase transition

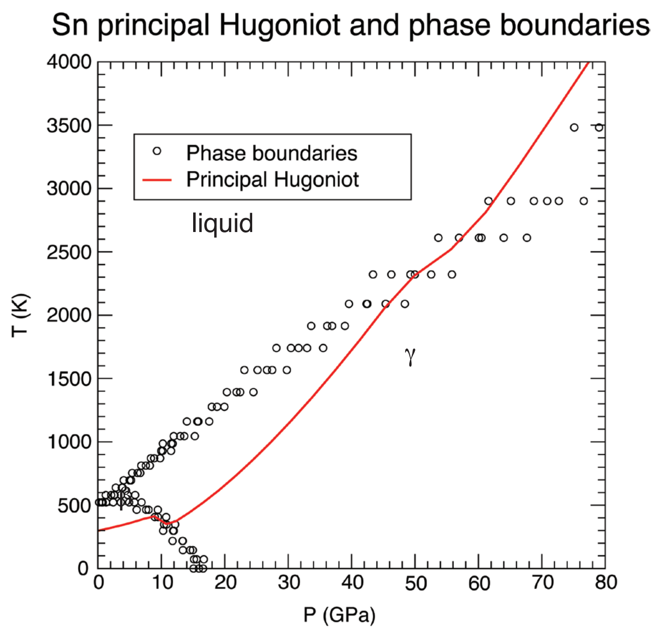

Fig. 2.
The principal Hugoniot of tin in pressure-temperature space, passing through all three phases. The effects of the phase transitions on the Hugoniot are clearly seen. Circles indicate the SESAME grid points nearest the phase boundaries.

is clearly visible in both the data and the EOS. The Hugoniot is shown over a larger range in Fig. 2, covering all three phases. The EOS is computed on a density-temperature grid, and grid points near the phase boundaries are indicated by circles.

We will continue to develop the code OpenSesame to address the new issues presented by this new EOS format, and we will also explore the challenge of extrapolating free energies well beyond the stability regions of the relevant phases, both of which will become even more important as we construct EOS for materials with more and more complicated phases.

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[1] E.D. Chisolm and D.C. Wallace, "Extending the CCW EOS II: Extending the Nuclear Contribution to High Temperatures," Los Alamos National Laboratory report LA-UR-04-3948 (June 2004).
[2] E.D. Chisolm and D.C. Wallace, "Dynamics of Monatomic Liquids," *J. Phys. Condens. Matter* **13**, R739 (2001).
[3] C.W. Greeff, et al., "Modeling Dynamic Phase Transitions in Ti and Zr," in *Shock Compression of Condensed Matter-2003*, M.D. Furnish, Y.M. Gupta, and J.W. Forbes, Eds. (Melville, NY: American Institute of Physics, 2004).